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Advances in magneto-ionic materials and perspectives for their application ⓔⓟ

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ABSTRACT

The possibility of tuning magnetic material properties by ionic means is exciting both for basic science and, especially in view of the excellent energy efficiency and room temperature operation, for potential applications. In this perspective, we shortly introduce the functionality of magneto-ionic materials and focus on important recent advances in this field. We present a comparative overview of state-of-the-art magneto-ionic materials considering the achieved magnetoelectric voltage coefficients for magnetization and coercivity and the demonstrated time scales for magneto-ionic switching. Furthermore, the application perspectives of magneto-ionic materials in data storage and computing, magnetic actuation, and sensing are evaluated. Finally, we propose potential research directions to push this field forward and tackle the challenges related to future applications.

I. MAGNETO-IONIC MATERIALS AS A NOVEL APPROACH TO ENERGY-EFFICIENT MAGNETIC SYSTEMS

Magnetic materials are important in a plethora of industrial applications, ranging from large-scale hard magnets in wind turbines and micromagnets in microelectromechanical systems (MEMS) to magnetic nanostructures for data storage and processing devices. Specific magnetic properties required for the individual applications are usually irreversibly set through the design of phase, composition, microstructure, and shape during the fabrication process. During the operation of magnetic devices, the direction of magnetization is conventionally controlled by the application of external magnetic fields, often realized by electromagnets or, in nanoscale devices, by large spin polarized electric currents. For both cases, Joule heating and associated energy dissipation present a severe challenge. This problem continuously triggers numerous research activities on magnetoelectric (ME) materials[1–3] in which the magnetic material is controlled by the electric voltage instead of electric current, thereby reducing energy consumption.

Magneto-ionic control of magnetic materials is a novel approach in the field of magnetoelectricity. The magnetic material in this case is in contact with a solid or liquid electrolyte. By the application of an external voltage, ionic motion and electrochemical reactions are triggered which can reversibly affect the performance of the magnetic material (Fig. 1). As the field is rapidly evolving, very diverse material/electrolyte systems are studied, and several denominations and categorizations, such as redox-based, electrochemical, ion-exchange, or magneto-ionic control of magnetism, are in use.[4–6] In this perspective, we consider voltage-tunable magnetic materials systems in which the underlying mechanism is based on ionic motion and electrochemical reactions as magneto-ionic materials. The main advantage of magneto-ionic materials, in contrast to most other magnetoelectric materials, is that they can be operated at room temperature and at low voltage. Since chemical changes are involved, the non-volatile setting of a magnetic state by voltage is possible, requiring only a small current. This is in contrast to the volatile magnetoelectric effects achieved by capacitive charging via the electrochemical double layer and associated surface electronic structure changes in similar gating architectures.[7,8] Thus,
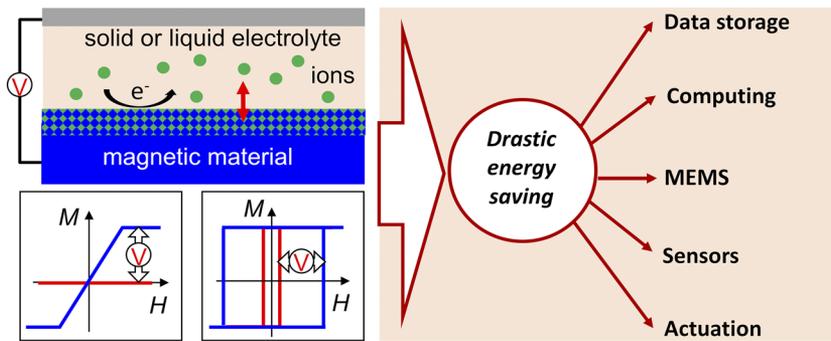
FIG. 1. Magneto-ionic systems consist of a magnetic material in contact with a solid or liquid electrolyte. The magnetic properties, such as saturation magnetization and coercivity, are controlled by voltage-triggered ionic motion and electrochemical reactions. This is promising for a drastic energy saving in comparison to conventional electric current-controlled magnetic devices in many possible applications.

magneto-ionic materials offer a route toward a reversible manipulation of magnetic properties and are highly promising for ultralow power magnetic devices.

The first reports on electrochemical voltage control of magnetism in ultrathin metal films dealt with oxygen-based interfacial reduction/oxidation and associated changes in magnetic anisotropy and coercivity.[9–13] Since then, numerous magneto-ionic material systems consisting of magnetic metals and/or oxides in combination with a solid or liquid electrolyte have been investigated; an overview of these advances until 2018/19 can be found in recent reviews on magnetoelectric materials.[4–6,14] Within the past two years, significant progress has been achieved, such as magneto-ionic materials that function on the base of proton[15–17] or nitrogen ion[18] transport, the extension toward 3D magneto-ionic systems,[19,20] and novel magneto-ionic functionalities such as tunable exchange bias.[21,22] In this expanding field, most reports still deal with the exploration of fundamental phenomena. With the key advantages of energy efficiency and room temperature operation, however, targeted research toward specific applications is expected in the future.

In this perspective, we give an overview on important latest advances in magneto-ionic research, discuss the challenges and opportunities of magneto-ionic materials, and propose future research strategies.

II. RECENT ADVANCES IN MAGNETO-IONIC RESEARCH

In the field of magneto-ionic materials, significant advances have been made within the past two years, especially regarding room temperature operation, switching speed, reversibility, and the extension of magneto-ionic tunability to diverse magnetic phenomena and functional materials. To illustrate the broad range of magneto-ionic materials, Fig. 2 gives recent examples for voltage-tunable magnetization curves in ultrathin single metal films[15] [Fig. 2(a)], metal alloy films[13] [Fig. 2(b)], oxide superlattices[17] [Fig. 2(c)], and three-dimensional oxyhydroxide nanoplatelet structures[19] [Fig. 2(d)].

In the following, we will focus on important advances from the past two years in light of the magnetic property, which is controlled by magneto-ionic means and with respect to application-relevant aspects, such as the switching speed and reversibility.

A. Magneto-ionic control of saturation magnetization

The voltage-control of the saturation magnetization (M_S) by magneto-ionic mechanisms has been demonstrated in a large variety of systems. To give an overview on the attainable M_S variations per defined voltage (V) change, the magnetoelectric (ME)-voltage coefficients, $\Delta M_S/|\Delta V|$,[4] are plotted in Fig. 3 for selected magneto-ionic systems and as a function of the time between the two
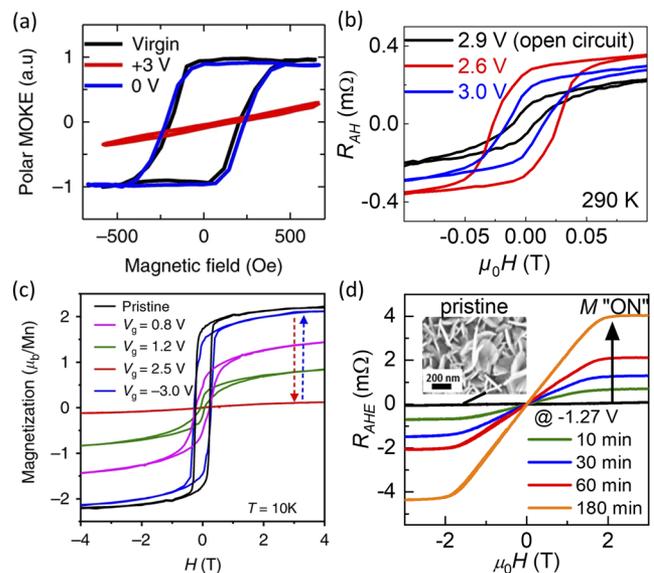
FIG. 2. Examples for voltage control of magnetic properties by ion migration and electrochemical reactions for various material systems. (a) Out-of-plane hysteresis loops corresponding to the virgin state and the first switching cycle for the proton-based magneto-ionic control in the Pt/Co(0.9 nm)/GdOx layer system. Reprinted with permission from Tan et al., Nat. Mater. **18**, 35 (2019). Copyright 2019 Springer Nature. (b) Out-of-plane hysteresis loops of an $L1_0$ CoPt film (2.8 nm) polarized in $LiClO_4$ in DMC/EC at different voltages (adapted from Reichel et al.[13]). (c) Out-of-plane hysteresis loops of a $[(La_{0.2}Sr_{0.8}MnO_3)_1(SrIrO_3)_1]_{20}$ superlattice under ionic liquid gating. Figure reproduced with permission from Yi et al., Nat. Commun. **11**, 902 (2020). Copyright 2020 Author(s), licensed under a Creative Commons Attribution 4.0 License. (d) Voltage-controlled ON-switching of magnetization by the electrochemical reduction of 3D FeOOH nanoplatelets in a 1M LiOH electrolyte (adapted from Nichterwitz et al.[19]).
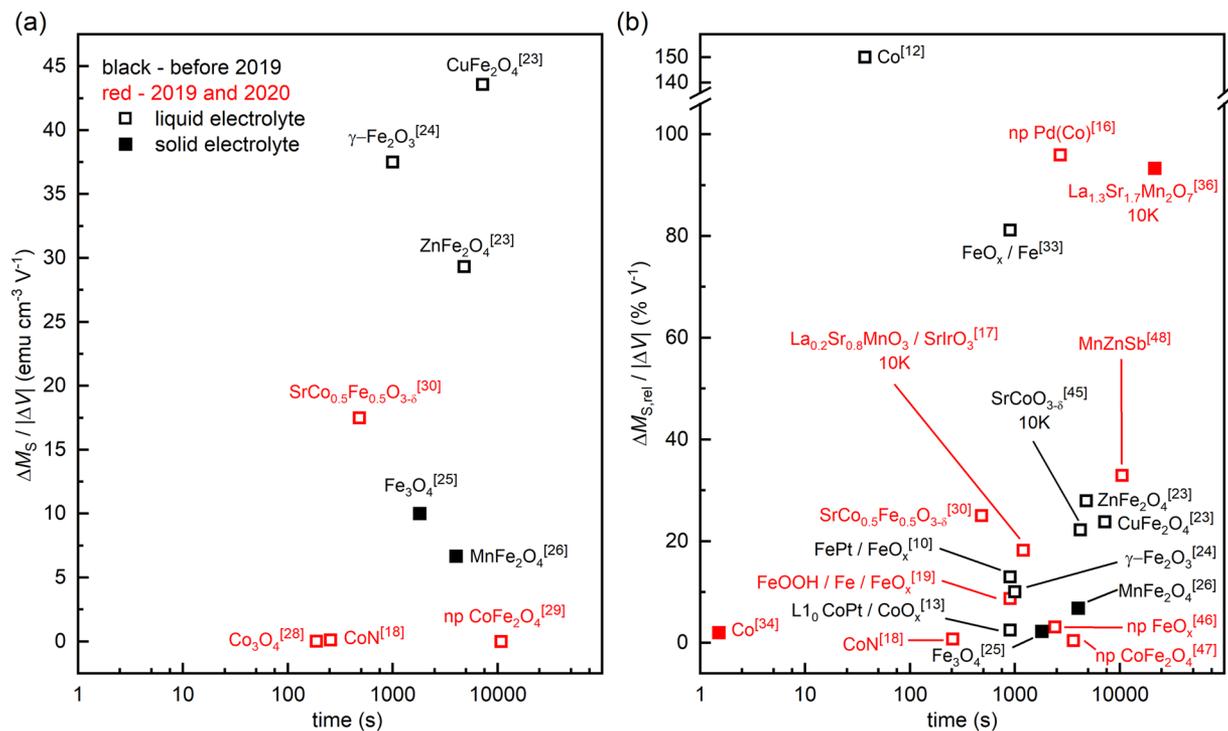
FIG. 3. Comparison of the ME-voltage coefficients for (a) an absolute and (b) relative variation in M_S for selected magneto-ionic systems as a function of the time elapsed between the setting of the two different M_S states considered in ΔM_S. The ME-voltage coefficients are given for the largest reversible M_S change. All data refer to room temperature measurements, except if otherwise stated. In (a), for reasons of comparability, only studies from which ΔM_S in emu cm^{-3} could be directly extracted are depicted. In (b), the maximum value of M_S is set as the reference value to ensure comparability: $\Delta M_{S,rel} = [M_{S1}(V_1) - M_{S2}(V_2)]/M_{S1}(V_1)$ with $M_{S1}(V_1) > M_{S2}(V_2)$. All data refer to room temperature measurements, if not stated differently [np—nanoporous].

voltage-induced M_S states. The data for very recent studies (from 2019 to 2020) are shown in red to highlight the current activities in the field. It should be noted that the time does not always represent the switching speed inherent to the material system, but sometimes also the time required for the magnetic measurement (which might be slower than the actual possible switching speed). Thus, the presentation in Fig. 3 gives the currently demonstrated time scales for magneto-ionic M_S-control.

Absolute values for the M_S variation per volt [Fig. 3(a)] are mainly reported for magneto-ionic systems based on ferrites and other transition metal oxides in thick film ($\geq$100 nm) or in bulk form.[23–30] Several systems, with both the liquid or solid electrolyte, are directly derived from battery research here and measured with *in situ* magnetometry.[23,25,26] In some cases, the magnetically active electrodes are prepared by mixing transition metal oxide nanoparticles with binders and conductive nanoparticles.[23,24,26,27] This procedure optimizes the electrochemical operation, but at the same time, the magnetic moment per volume is diluted. Figure 3(a) shows that, so far, the largest ME-voltage coefficients for M_S are in the range of several tens of emu cm^{-3}/volt. This seems small in comparison to conventional magnetic materials exhibiting high M_S (e.g., CoFe alloy with M_S up to 1950 emu cm^{-3}); however, in the studied systems with small overall M_S, the magneto-ionic effects are sufficient

to modulate M_S to a large extent. For instance, a 70% decrease in M_S is reversibly achieved by Li intercalation in $ZnFe_2O_4$ for an applied voltage of just 1.25 V.[23] For most systems in Fig. 3(a), the time scales for the magneto-ionic measurements range from minutes to hours, which is very long with regard to applications. However, it must be acknowledged that most studies focused on the fundamental understanding of the mechanisms, targeted large tunable volumes, and utilized time-intensive measurements such as *in situ* superconducting quantum interference device (SQUID) magnetometers.[26,27]

Recently, Ning *et al.*[30] reported non-volatile magneto-ionic control of M_S in epitaxial $SrCo_{0.5}Fe_{0.5}O_{3-\delta}$ (SCFO) thin films via ionic liquid gating. Reversible and continuous control of the magnetization up to 100 emu cm^{-3} was demonstrated at room temperature, which was ascribed to voltage-induced changes in the oxygen stoichiometry. At negative voltage (-2 V), oxygen is extracted from the perovskite SCFO, which leads to the decrease in the magnetic moment (80% decrease in M_S after 2 min). After prolonged gating, the nonmagnetic brownmillerite phase forms and thus ferromagnetic ordering is suppressed ($M_S \sim 0$ after 3 min). The ferromagnetic state can be recovered by applying a positive voltage ($+2$ V) at which oxygen is reinserted. In this way, reversible ON/OFF control of M_S is obtained by alternating the polarity of the gating voltage. The switching speed of few minutes is

remarkable, especially in view of the relatively large film thickness of up to 150 nm.

De Rojas et al.[18] proposed the use of nitrogen ions instead of oxygen ions to improve the magneto-ionic switching speed. They demonstrate a reversible ΔM_S of 1 emu cm^{-3} upon the electrolytic gating of CoN (85 nm) thin films in propylene carbonate (PC) with solvated Na$^+$ species. To reach a ΔM_S of 1 emu cm^{-3}, in this system, voltages of ± 4 V and switching times around 4 min are required. A higher voltage of ± 8 V and a longer time of 6 min are required to achieve the same effect in Co$_3$O$_4$ films of the same thickness. This difference between the magneto-ionic behavior of CoN and Co$_3$O$_4$ is discussed with regard to the lower activation energy for ion diffusion and the lower electronegativity of nitrogen compared to oxygen. In a further study, de Rojas et al.[28] revealed the importance of the gating geometry for the magneto-ionic efficiency for Co$_3$O$_4$. The insertion of a conductive buffer layer underneath the semiconducting and magneto-ionically active Co$_3$O$_4$ film (130 nm) to realize a bottom gate electrode yields faster switching and an increase in the ME-voltage coefficients from around 0.01 emu cm^{-3} V^{-1} to 0.11 emu cm^{-3} V^{-1}.[28] For the magneto-ionic control of Co$_3$O$_4$[28] and CoN,[18] in comparison to Li intercalation in spinel ferrite systems,[23] the ME-voltage coefficients are significantly smaller, but faster switching is demonstrated.

The relatively scarce data for absolute ME-voltage coefficients (with ΔM_S in emu cm^{-3}) in Fig. 3(a) are connected to measurement limitations for magneto-ionic materials, as they are often utilized in the ultrathin film or nanoporous form. One challenge is the difficulty to accurately determine the volume of these materials. A second challenge is that the very small magnetic moments, as in ultrathin films, are usually not measurable with conventional magnetometry, especially if in situ characterization requires further addition of the electrolyte and cell components. Therefore, for ultrathin films, surface-sensitive magnetic techniques based on the anomalous Hall effect (AHE) or the magneto-optical Kerr effect (MOKE) are utilized to probe relative changes in magnetization during magneto-ionic switching.[15,31,32] To give a more comprehensive overview, ME-voltage coefficients for the relative variation in M_S over time are plotted in Fig. 3(b). Here, the great variety and extended activities regarding magneto-ionic material systems become obvious. Effective magneto-ionic control of M_S can also be achieved in systems containing metallic ferromagnets, such as Fe,[33] Co,[34] FePt,[10] CoPt,[13] and Pd(Co).[16]

In the FeO$_x$/Fe system, the magneto-ionic switching relies on the reversible electrochemical transformation (surface oxidation/reduction) between weakly magnetic iron oxide and ferromagnetic iron metal in alkaline aqueous solution.[35] In this oxygen-ion based system, Duschek et al. achieved relative variations of M_S up to 64% in continuous FeO$_x$/Fe films (2 nm)[32] and almost ON/OFF switching of M_S in FeO$_x$/Fe nanoisland geometry[33] for a voltage change of just 1 V. The increased magneto-ionic effect in FeO$_x$/Fe nanoislands, as compared to the thin film geometry, is attributed to the larger surface/volume ratio, which promotes the interfacial oxidation/reduction reactions. Recently, a continuous voltage-triggered ON switching of magnetism based on the reduction of paramagnetic β-FeOOH nanoplatelets to ferromagnetic Fe [Fig. 2(d)] has been achieved in the same electrolyte.[19]

The insertion and removal of oxygen is also proposed as the origin for the voltage-induced change in M_S in Co films (0.8 nm) gated via a GdO$_x$ layer.[34] Here, voltage pulses of ± 10 V were applied and continuous changes in magnetization were recorded down to a time scale of milliseconds. Di et al.[12] also reported variations of M_S (up to 60% M_S reduction) of an ultrathin Co layer. The effects were attributed to voltage-triggered reversible Co surface oxidation in the liquid alkaline electrolyte. The effects occur within a very small voltage range (ΔV of 0.4 V) and are demonstrated in a time scale of about 40 s.

For nanoporous Pd(Co) polarized in 1M KOH solution, Gößler et al.[16] demonstrated a fully reversible voltage-induced ON and OFF switching of magnetization for a voltage change of about 1 V. This strong magneto-ionic effect is discussed with regard to electrochemical hydrogen sorption, which affects the magnetic coupling of the Co clusters via the Ruderman–Kittel–Kasuya–Yoshida-type interaction in the Pd matrix. In consequence, magneto-ionic switching between the ferromagnetic and the superparamagnetic state is achieved.

There have been remarkable advances also with regard to the voltage-control of magnetization in perovskite transition metal oxides by ionic mechanisms.[17,36,37] In superlattices comprised of alternating one unit cell of SrIrO$_3$ and La$_{0.2}$Sr$_{0.8}$MnO$_3$, protons and oxygen ions can be transferred using ionic liquid gating, which triggers reversible phase transitions.[17] As a result, voltage-controlled ON/OFF switching of M_S [Fig. 2(c)] is demonstrated. Vasala et al.[36] studied the electrochemical (de)intercalation of fluoride ions into a two-layer La$_{1.3}$Sr$_{1.7}$Mn$_2$O$_7$ system to tune the magnetization by exploiting the high sensitivity of the magnetic states to the Mn oxidation state and the distance between the perovskite building blocks. They achieve a 67% change in M_S for low applied voltages (<1 V), which is reflected in a high ME-voltage coefficient [Fig. 3(b)]. So far, the large magneto-ionic effects are restricted to a low temperature (e.g., 10 K) here.

B. Magneto-ionic control of magnetic anisotropy and coercivity

The control over magnetic anisotropy and coercivity (H_C) is a key requirement for the design of magnetic materials in most types of applications. Figure 4 gives an overview on ME-voltage coefficients for H_C achieved by magneto-ionic mechanisms, again as a function of the time passed between the settings of the two states. Most studies deal with ultrathin Co films exhibiting perpendicular magnetic anisotropy (PMA).[9,11,12,15,38] In such films, the anisotropy and spin reorientation can be controlled by magneto-ionic mechanisms, resulting in an ON/OFF-switchable coercivity and remanence [Fig. 2(a)]. Recently, large magneto-ionic effects on anisotropy and H_C have also been reported for Fe films with in-plane uniaxial anisotropy[39] and for typical hard magnetic materials such as L1$_0$ CoPt[13] and SmCo$_5$.[40] This is a promising sign that magneto-ionic control can be transferred to a wide range of usable magnetic materials in the future.

For the magneto-ionic control of anisotropy switching in ultrathin Co layers voltage-gated via GdO$_x$, advances in switching speed and reversibility are notable. Tan et al.[15] demonstrated the benefits of protons over oxygen ions as the functional ions and achieved fast (100 ms) and highly reversible (2000 cycles) anisotropy switching at room temperature. In previously utilized GdO$_x$/Co structures, which relied on oxygen ion migration, significant magneto-ionic
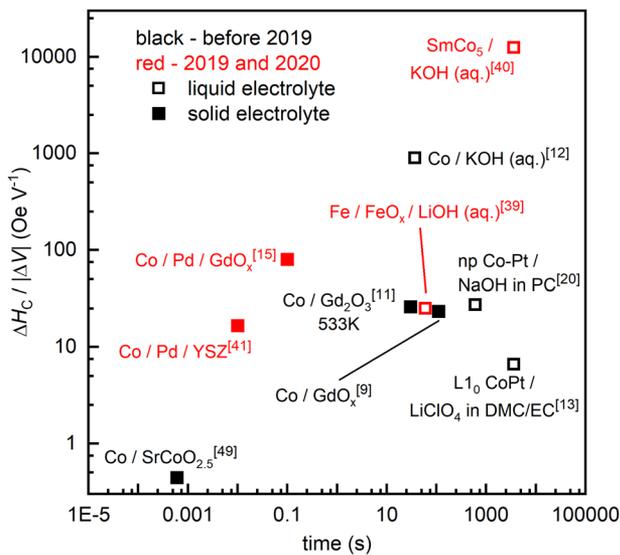
FIG. 4. Comparison of the ME-voltage coefficients for variation in H_C for selected magneto-ionic systems as a function of the time elapsed between the settings of the two different H_C states considered. The ME-voltage coefficients are given for the largest reversible H_C change. All data refer to room temperature measurements, if not stated different (aq.—aqueous solution, DMC/EC—dimethyl carbonate and ethylene carbonate, PC—propylene carbonate, YSZ—yttria-stabilized zirconia, and np—nanoporous).

compared different proton conducting gate oxides and their effect on the switching speed for the magneto-ionic control of Co layers. The remanence ratio approaches zero within a time scale of >80 s for gadolinium doped ceria (GDC) and barium cerium yttrium zirconate (BZCY), while it takes less than 1 s for yttria-stabilized zirconia (YSZ). The Pt/Co/YSZ/Au structure is further optimized toward a Pd/Co/Pd/YSZ/Pt architecture in which Pt is used as top electrode to catalyze the electrochemical reaction and Pd serves as a hydrogen loading layer as well as a protection layer between the YSZ and the Co. Here, the fastest switching speed of <2 ms is achieved for a thickness of 10 nm for YSZ and with a gate voltage of +6 V. Huang et al.[42] recently showed that voltage-tunable PMA on the base of oxygen ion migration can also be achieved in $Pt/Co/CoO/TiO_2(TaO_x)$ heterojunctions. In combination with the spin current generated from the Pt layer, this makes voltage control of the spin-orbit torque (SOT) induced perpendicular magnetization switching of Co possible.

Besides the substantial work on magneto-ionic control of ultra-thin Co films with PMA, an extension to films with in-plane anisotropy has recently been reported. Zehner et al.[39] achieved a fully reversible low voltage-induced collapse of coercivity and remanence in FeO_x/Fe films (10 nm) with uniaxial in-plane anisotropy during electrolytic gating at low voltage in 1M LiOH aqueous solution [Figs. 5(a) and 5(b)]. In the initial FeO_x/Fe films, Néel wall interactions stabilize a blocked state with high coercivity. With dedicated and angle-resolved in situ Kerr microscopy, working in combination with liquid electrolyte gating, inverse changes in coercivity and anisotropy and a coarsening of the magnetic microstructure [Fig. 5(c)] were probed. The quantitative analysis of the anisotropy and domain size changes allowed us to reveal the redox-induced change in Néel domain wall interactions and in the microstructural domain wall-pinning sites as the origin for the voltage-triggered magnetic deblocking. This reversible modulation of local defects to tune H_C goes beyond state-of-the-art thin film magneto-ionics in which extrinsic properties such as coercivity are usually changed by controlling the intrinsic magnetic anisotropy. With this approach, voltage-assisted 180° magnetization switching with a high energy efficiency is achieved within seconds. Zehner et al.[21] further extended the magneto-ionic functionality to

effects required high temperatures or were restricted to the edges of the patterned electrodes.[9] For the proton-based mechanism, the authors suggested that protons, generated by atmospheric water splitting at the Au top electrode, are transported through GdO_x to the Co interface.[15] For this mechanism, a sufficiently humid atmosphere is required. The magnetic anisotropy switching is ascribed to the high sensitivity of the interface anisotropy of Co to adsorbed H. In the same work, anisotropy switching is also observed for a $Au/GdO_x/Pd/Co/Pd$ structure, which is in this case explained by the hydrogenation of the Pd layer adjacent to the Co layer. Lee et al.[41]

FIG. 5. Voltage-control of coercivity and magnetic domains in FeO_x/Fe films with uniaxial in-plane anisotropy. (a) In-plane hard axis magnetization curve in pristine state and at -0.02 V (oxidation) and at -1.1 V (reduction) showing the voltage-induced collapse of coercivity and remanence. (b) Angular dependence of coercivity for the pristine (=oxidized) and the reduced state. (c) Magnetic domains in reduced and oxidized state as observed by in situ Kerr microscopy (adapted from Zehner et al.[39]).

in-plane exchange bias thin films, which are important for stable and artificial magnetization distributions.[43] The authors could demonstrate voltage-controlled non-volatile programming of exchange bias fields in $FeO_x/Fe/IrMn$ heterostructures with fully shifted hysteresis loops by a redox-based transition of the ferromagnetic layer.

Recent advances are reported also for the magneto-ionic control of coercivity and magnetization in bulk-like materials. Navarro-Senent et al.[20] studied mesoporous Co–Pt/CoO microdisks (>300 nm) in PC with traces of NaOH. The voltage-driven oxygen migration promoted a partial reduction of CoO to Co and led to a strong decrease in coercivity (−85%) at −10 V, which could be partially recovered. In a subsequent study, Navarro-Senent et al.[44] investigated the behavior of nanoporous Co–Pt films covered by 10 nm AlO_x or HfO_x in the same electrolyte. Differences in the magneto-ionic behavior of uncoated Co–Pt films are ascribed to the oxygen acceptor behavior of HfO_x and the oxygen donor behavior of AlO_x.

For micrometer-sized $SmCo_5$, Ye et al.[40] showed a huge modulation of coercivity (ΔH_C of 1 T for $\Delta V < 1$ V) by electrochemical hydrogenation in 1M KOH aqueous electrolyte. Even though very long timkes are required for the largest H_C change, voltage-assisted magnetization reversal could be demonstrated within around 3 min. Further improvements in switching speed are proposed by the reduction of the particle size, the introduction of additional diffusion paths, or optimized device geometries.

III. APPLICATION PERSPECTIVES FOR MAGNETO-IONIC MATERIALS

In the following, application perspectives for magneto-ionic materials in the fields of memory devices, neuromorphic computing, spin-torque nano-antennas, sensors, and magnetic actuation are discussed.

Digital memory is a crucial part in our current digital era. The advent of internet of things, machine learning, and artificial-intelligence (AI) based applications, such as image recognition[50] or protein folding,[51] impose strict requirements on the processing speed and energy efficiency of data-centric tasks. Already today, the worldwide energy consumption of the data centers of all cloud service providers is about 200 TW/year.[52] Magnetic hard disk drives (HDD) remain a conventional way to store data in computers and large data centers, but the electric currents required for data writing are associated with the energy loss due to Joule heating. Also, charge based memories, such as flash and dynamic/static random-access memory (RAM), have high energy consumption and further device miniaturization becomes problematic mainly because of increasing leakage currents.[53] The use of magnetic elements for RAM seems appealing due to non-volatility and fabrication compatibility with CMOS.[54] Additionally, complex magnetic textures can be used for information storage.[55,56] In such magnetic memory concepts, electric currents are still used to control magnetization dynamics. The combination of magnetic memory concepts with energy-efficient magneto-ionic control could be a route to reduce the operational energies for next generation data storage media.

For HDD, control of coercivity or magnetization of the storage media by a voltage, instead of a magnetic field, is appealing to reduce the energy consumption. In high anisotropy magnetic thin films, which are required for data stability in high density

magnetic data storage, a temporary reduction of coercivity during the writing process (magnetization switching) is desirable. So far, this is accomplished by heat-assisted magnetic recording,[57] which, however, involves the generation of heat with a laser. Here, provided that the switching speed is improved, magneto-ionic mechanisms to control coercivity may be a route to more energy-efficient voltage-assisted magnetic recording.

The device structure and nanoscale ion transportation mechanism in some magneto-ionic devices are comparable with those of resistive switching devices, which are used for the non-volatile resistive random access memory (RRAM).[58] It has been experimentally demonstrated that device resistance can be switched among multistates to achieve an ultra-high density/capacity storage.[59] In analogy, non-volatile ion transportation triggered by voltage could result in progressive changes in the magnetization value in magneto-ionic materials. Thus, we propose that the stable multi-level intermediate states observed in many magneto-ionic materials could be extended into a multi-state magnetic storage concept.

Current-driven magnetic domain wall motion has raised hopes for new memory or logic devices, but so far, high threshold current densities and defect pinning effects pose challenges to the further development of this concept.[60] Magneto-ionic approaches have already shown that domain-wall propagation fields can be tuned through oxygen ion-based magneto-ionic control of ultrathin Co layers[61] and Fe films.[39] Offering the advantage of lower threshold current densities and topologically protection,[62] skyrmions are an alternative candidate to domain wall memory logic devices. One way to stabilize skyrmions is via the Dzyaloshinskii–Moriya interaction (DMI). By adjusting the strength of the DMI, it is possible to tune the domain wall chirality and the skyrmion's winding number, making it more suitable for practical applications. Interestingly, with regard to potential magneto-ionic control, DMI can be induced by oxygen chemisorption on a ferromagnetic surface.[63] A recent work demonstrated that the DMI strength can be (so far irreversibly) tuned by oxygen-ion migration at the ferromagnet/heavy metal (Co/Pt) interface.[31] Furthermore, gas phase experiments have shown that skyrmions can be induced in ultrathin Fe upon hydrogen exposure.[64] This indicates that modulation of skyrmions by fast proton-based magneto-ionic control may be feasible.[15] Fine tuning and reversibility of skyrmion parameters with high endurance, for example, through hydrogenation, could be favorable for the next generation spin-orbitronic devices.

In general, the limiting factors for future applicability in data technology might be, in the first place, the reversibility and switching speeds. In order to compete with the state-of-the-art technology, a stability over >10^{15} cycles and switching speeds between 10 ms (HDD) and 10 ns [spin transfer torque (STT) RAM] are desirable.[65]

The practical applications of AI require processing of large volumes of data, which, in conventional computing architecture (separate processing and storage units), is energy expensive and limited by von Neumann bottleneck and the memory wall.[66] Neuro-inspired computing chips that emulate the working principles of the biological brain are expected to perform better and more energy efficient.[67] Memristive elements are shown to emulate synaptic properties.[68] Since latest magneto-ionic approaches resemble the redox memristors in its operational principles, they may serve as an alternative building block for neuromorphic hardware. An artificial synapse and

different synaptic functions based on the magneto-ionic setting of different magnetization states were already demonstrated in the Co thin film.[34]

Magnonics is gaining a lot of popularity for wave-based computing[69] and microwave electronic[70] applications. It has been predicted that terahertz (THz) spin waves can be excited by domain wall motion induced by STT and SOT.[71] Large bandwidths of frequency modulation can be achieved, which is important for signal processing[72] and in telecommunication technologies. It was theoretically demonstrated in antiferromagnets that the wavelength of the emitted spin waves depends on material lattice constants.[73] One of the limitations to excite the spin waves is the high threshold current density in materials with strong exchange interactions. Here, the magneto-ionic mechanism such as ion intercalation or hydrogenation may be an approach to tune the lattice constants and the exchange interactions to obtain the desired wavelength.

Spin-torque nano-oscillators are magnetic tunnel junctions that emit microwave signals when a direct current induces sustained magnetization precession. A frequency tuning of 50 MHz has already been demonstrated in an electrically gated spin Hall nano-oscillator by engineering the electronic bands at the ferromagnetic/oxide interface.[74] However, an all ionic control of PMA would potentially lead to higher modulation of damping,[75] thereby directly influencing the auto-oscillation threshold current.

The opportunities to tune magnetic properties by a low voltage are also of interest for magnetic sensors. Modern magnetic field sensors rely on the giant magnetoresistance (GMR) or tunneling magnetoresistance (TMR) effect in functional layer stacks and are key to the information technology and numerous scientific and industrial measurement applications. Voltage control of GMR and TMR stacks via the electric field control of the magnetic anisotropy is proposed as a promising route to tuning and enhancing the sensitivity and the linear range of magnetic sensors.[76,77] With magneto-ionic approaches, ultralow voltage and non-volatile programming of magnetic sensors may be within reach, but this remains largely unexplored. One recent study on oxide/metal layers demonstrates a tunable magnetoresistance (MR) and even a voltage-controlled sign change in MR by magneto-ionic mechanisms.[78]

A different type of potential sensor application relates to the interplay of electrochemistry and magnetism, which is inherent to magneto-ionic materials. The magnetic state not only depends on the applied voltage but also on the type of electrolyte species, pH, and hydration state, and thus, it can be regarded as a magnetic fingerprint to sense solution, solid-state, or atmospheric chemistry. Such a direct magneto-chemical sensor would have the advantage that functionalized magnetic particles, which are often utilized in magneto-chemical sensing platforms,[79] are not required. A first example in this direction is *in situ* characterization of (de)lithiation processes in Li_xCoO_2 cathodes for Li ion batteries by utilizing *in situ* SQUID magnetometry.[80]

Magnetic actuation is important in a plethora of modern technologies, including microfluidic chips,[81,82] (micro)robotics,[83] and MEMS.[84] Up to now, the magnetic actuation functionality requires an external magnetic field as the control parameter, which is usually provided by an electromagnet. This becomes problematic particularly when downscaling to the microscale, as here, high current densities are required to achieve sufficient magnetic field strength.[84] Switchable micromagnets by magneto-ionic control could present

an energy-efficient alternative to standard micro-electromagnets, especially as the demand for high switching speed is less crucial than for memory devices. As an example, in some magnetophoretic devices, particle transport in a liquid above a magnetic film is realized by magnetic actuation in a time scale of seconds via the modulation of artificial magnetic domain patterns.[85] The required external magnetic field is so far realized by electromagnets. Direct magneto-ionic control of the magnetic domains[21] could potentially replace magnetic field control in such devices in a succinct way.

To achieve sufficient stray fields for actuation, a high energy product of the magnet is required. Thus, materials with high saturation magnetization, as a precondition for a high remanence, and sufficient H_C will be favorable here. Furthermore, magneto-ionic materials exhibit the unique feature of different magnetic states (H_C, remanence) being able to be set by an external voltage, and this opens the door to a voltage-programmable actuation functionality.

IV. POTENTIAL RESEARCH DIRECTIONS AND CONCLUDING REMARKS

While the technology readiness of this emerging topic is still at the fundamental research levels 1 and 2 (1—basic principles observed and 2—technology concept formulated), the recent research works clearly show that magneto-ionic control can be extended to a broad range of material systems and magnetic functionalities. In the following, we propose potential research directions to push this field forward, especially in view of potential applications.

So far, magneto-ionic control is demonstrated for academic material systems and the material choice has primarily been conducted on the base of the targeted magneto-ionic mechanism. Now, with the basic knowledge on magneto-ionic principles, the research could extend toward magnetic materials, which exhibit optimized initial magnetic properties with regard to application. Here, research can rely on long-standing engineering approaches to design magnetic materials, including the optimization of the microstructure, texture, intergranular phases, or composition in alloys. For example, magneto-ionic control of bulk ferrites and rare earth hard magnets is so far restricted to isotropic materials.[29,40] An extension to textured materials could push forward the application potential for actuation, as the accessible energy product could be increased. Furthermore, the search for magneto-ionic mechanisms to control antiferromagnetic layers, which are currently hot candidates for spintronic devices, seems promising. In this regard, Mustafa *et al.*[22] recently demonstrated voltage control of exchange bias by the intercalation of Li in antiferromagnetic Co_3O_4, placed adjacent to the ferromagnetic Co layer.

The energy efficiency of magneto-ionic systems is the most intriguing aspect for their application potential. Research works on magneto-ionic control already clearly demonstrate that a large variety of magnetic properties can be tuned by a low voltage. The electrochemical reactions involve electric currents, but these are orders of magnitude smaller than those required for electric current controlled magnetic devices. In addition, as the mechanism relies on chemical changes, the effects are often non-volatile and thus do not require a continuous power supply. For the FeO_x/Fe system, magneto-ionic switching energies in the range of few femtojoules (for a 50 nm device) are projected,[39] which is comparable to the lowest reported switching energies for magnetic

tunnel junction devices.[86] Considering that the switching speed is not yet optimized, this indicates that magneto-ionic mechanisms can yield an improvement in the energy efficiency of several orders of magnitude. Nevertheless, quantitative information on the energy consumption of magneto-ionic systems is still too scarce and should be provided more often in the future to highlight the competitiveness with alternative approaches in magnetic technologies.

The limitation of the switching speed, due to the required ionic motion, is considered as one of the main challenges for the application of magneto-ionic materials. Consequently, future research should address this issue. For ultrathin Co films with PMA, which is the most-studied magneto-ionic material, the optimization of the device architecture, film thickness, and gate oxide has successfully increased the switching speed to the range of (sub)milliseconds[41] and, for small effects, also to the sub-nanosecond regime.[49] With this, these systems approach the required switching speeds for memory applications. Only for few systems besides Co, e.g., for the FeO_x/Fe system, a switching speed in the range of seconds is reported,[39] which may suffice for actuation and sensor applications. Time scales of hours to days, which occur for many magneto-ionic systems (see Figs. 3 and 4), seem unacceptable from an application standpoint. In many cases, actually, the maximum possible switching speed may not be known due to measurement restrictions. Thus, a deeper understanding and an improvement of the key aspects for the switching speed is an important research strategy. In a first step, time-resolved magnetic measurements would offer access to the "true" switching speed. Second, interface- and defect-engineering could be pursued as a promising route to optimize the ion transport pathways and thus the kinetics, similar to the successfully applied concepts in the field of memristive systems.[87]

The reversibility of magneto-ionic effects is a further important issue that defines the possible application areas. At first glance, magneto-ionic systems without a structural phase transformation (such as Li intercalation in spinel structure[24]) or with purely interface effects (such as surface effects at ultrathin Co films[12,15]) seem favorable here. In contrast, many magneto-ionic systems involve a phase transformation, which exponentiates tuning possibilities but may decrease reversibility due to strain-related fatigue or irreversible microstructure changes. In such systems, good reversibility is often found at the surface/interface regions or for nanoparticulate morphologies.[28,33,88,89] Thus, the minimization of the affected volume, e.g., via interface functionalization in nanostructured materials, seems to be a promising route to high reversibility. Furthermore, the unprecedented possibilities of magneto-ionic fine tuning of material properties by voltage at room temperature, even in the case of limited reversibility, should be acknowledged as a novel material design approach. Indeed, non-volatile magneto-ionic manipulation has already been proposed as the energy-efficient patterning method for artificial domains in exchange bias systems[21] or to set specific DMI values.[31]

Magneto-ionic systems often involve liquid electrolytes. There are several advantages of liquid electrolytes over solid electrolytes, namely, the high electric fields that are accessible due to electrochemical double layer formation, the possibility to infiltrate 3D geometries, the easier characterization of solid/liquid in comparison to solid/solid interfaces, and the more straightforward understanding of mechanisms in the frame of conventional electrochemistry

models. At the same time, the presence of a functional liquid is a critical point for implementation in most technologies. Therefore, research efforts should also target a transfer from solid/liquid systems to all-solid-state magneto-ionic systems wherever possible. Recent examples are the use of ionic gels,[90] hydrated oxides,[15] and devices that are derived from solid state battery architectures.[25,36] On a more futuristic level, solid/liquid magneto-ionic principles may become useful in brain-inspired computing concepts[91] or for magnetic actuation in microfluidic devices.[85]

So far, research on magneto-ionic materials focused, to a large extent, on the basic science. Thus, in order to move from academic research to the application of magneto-ionic materials, the development of various laboratory demonstration prototypes is an essential future task.

In summary, this perspective gives an overview about important research advances in the field of magneto-ionic control of magnetism. These include the growing diversity of magneto-ionic materials systems and functionalities and the improvements toward high switching speeds. We discuss various application perspectives for magneto-ionic materials in memory, computing, actuation, and sensor technologies and suggest potential research directions. For instance, the quantification of switching energy and switching speeds and the development of magneto-ionic prototype systems seem important to highlight the energy saving potential and increase the competitiveness for future applications.

AUTHORS' CONTRIBUTIONS

M.N. and S.H. contributed equally to this work.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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